

## Permittivity in the microwave region of some silicate and ferrite minerals at low pressures

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The dielectric permittivity and attenuation of some silicate and ferrite minerals have been measured at very low pressures. The measurements are carried out in the microwave region at 8.63 GHz using a standing wave method. Both dielectric permittivity and attenuation caused by the inserted sample are increased by increasing the pressure. The results obtained have been interpreted.

### 1. THEORETICAL

The permittivity and dielectric loss in the microwave region can be determined by several methods (Comer & Smyth 1943, Jackson 1946, Jackson & Powles 1946, Penrose 1946, Roberts & Von Hippel 1946, Heston *et al* 1948). In this work a method (Roberts & Von Hippel 1946) which depends on standing wave measurements in a rectangular waveguide is used.

Let the waves propagating from the source be incident normally at the surface of the material of dielectric constant  $\epsilon^*$ . The reflection coefficient  $R$  is given by  $R = |R| e^{i\phi}$ , where  $\phi$  is the phase change on reflection. As a result of reflection, a standing wave pattern is produced and from the shape and position of this pattern  $\phi$  and  $|R|$  can be obtained.

From the transmission line theory,  $R = \frac{1-S}{1+S} e^{i\theta}$  where  $S$  is the reciprocal of the standing wave ratio. If  $S_0$  and  $S_S$  are the reciprocal of the standing wave ratio in the open and short circuited cases, the reflection coefficient  $|R|$  for the two previous cases denoted by  $|R_0|$  and  $|R_s|$  can be calculated.

According to Heath (1962), the attenuation ( $\alpha$ ) in decibels for very low loss materials can be calculated from

$$\alpha = 10 \log \frac{|R_0| + |R_s|}{2} \quad (2)$$

From the standing wave ratio and phase shift, the input impedances  $z_0$  and  $z_s$  in the open and short circuited cases can be calculated using the Smith Chart.

and used to find  $\epsilon'$  and  $\epsilon''$  (Hanna & Bishai 1966) from

$$\frac{1}{r_0 r_s} = \frac{\epsilon' - q}{1 - q} - i \frac{\epsilon''}{1 - q}$$

where  $q = (\lambda/\lambda_c)^2$ ,  $\lambda$  is the wavelength in air and  $\lambda_c$  is the cut-off wavelength.  $\epsilon'$  and  $\epsilon''$  can be obtained by equating the real and imaginary parts of the last equation. The values obtained are the measured values and can be denoted by  $\epsilon'_m$  and  $\epsilon''_m$  and can be used to calculate  $\epsilon'$  and  $\epsilon''$  of the material according to Balls (1946), Kirkwood *et al* (1954) and Ishida *et al* (1959),

$$\epsilon'_m = 1 + P(\epsilon' - 1)$$

$$\text{and} \quad \epsilon'' = \frac{\epsilon''_m}{P}, \quad \dots \quad (3)$$

where  $P$  is the density of packing of the material as given by the ratio of the volume of the material to the overall volume occupied. The volume of the material is obtained from the mass divided by density and the overall volume occupied from the dimensions of the space occupied by the material. The accuracy in  $\epsilon'$  is better than 5%.

## 2. EXPERIMENTAL

The apparatus used for the measurements of permittivity and attenuation caused by the inserted samples in the microwave region is described by Hanna & Bishai (1966). The cell used in all the measurements is a piece of waveguide 4 cm long and closed from both sides by thin mica windows. The cell is closed by a shorting metal plate for the measurements in the short circuited case. For the measurements in the open circuited case, the cell is connected to a movable short adjusted to interpose a  $\lambda/4$  air filled waveguide between it and the back face of the test material.

Measurements were carried out on clinocstatite, forsterite, magnesioferrite, magnesiochromite,  $\beta$ -dicalcium silicate and tricalcium silicate. Each sample was dried at 200°C for six hours till a constant weight is attained before use and compressed inside the cell, by means of a plunger and an oil press. The voltage standing wave ratio is determined by taking the difference  $\Delta_x$  between the probe positions at which the power equals twice its minimum value and substituting in the equation (Ginzton 1957),

$$S = \frac{1}{\sigma} = \frac{\pi \Delta x}{\lambda g}$$

Also, from the position of the first minimum from the interface,  $\phi$  can be computed. Knowing  $S_0$ ,  $S_s$  and  $\phi$  the values of  $r_0$  and  $r_s$  can be obtained and used to find  $\epsilon'$  and  $\epsilon''$ .

### 3. MATERIALS

#### *Clinoenstatite and forsterite minerals:*

Clinoenstatite and forsterite minerals are characterised by their good dielectric properties and have promoted their extensive use in the production of insulators for high frequency specimens. Standard clinoenstatite ( $\text{MgO} \cdot \text{SiO}_2$ ) minerals were prepared synthetically from finely ground mixtures of MgO and silica in the molecular proportion of 1:1 and 2:1, respectively. The homogeneous mixtures were fired on two periods of 10 hours at  $1350^\circ\text{C}$  in case of clinoenstatite minerals and  $1600^\circ\text{C}$  in case of forsterite minerals with intermediate grinding.

#### *Magnesio-ferrite and magnesiochromite spinel minerals:*

Magnesioferrite ( $\text{MgO} \cdot \text{Fe}_2\text{O}_3$ ) and magnesiochromite ( $\text{MgO} \cdot \text{Cr}_2\text{O}_3$ ) spinels are important in high frequency engineering. They play a role as materials with high electric resistance.

Standard magnesioferrite spinel was obtained synthetically from the finely ground mixture of MgO and  $\text{Fe}_2\text{O}_3$  in a stoichiometric ratio. Homogenization of the mixture in a special steel-bull-mill was carried out for a period of 6 hours followed by firing to the sintering point at temperatures ranging between  $1000$ – $1300^\circ\text{C}$  in the presence of a stream of oxygen.

Standard magnesiochromite was prepared synthetically from a mixture of finely ground MgO and  $\text{Cr}_2\text{O}_3$  in the molar-ratio 1:1. The presence of 0.5–1% excess  $\text{Cr}_2\text{O}_3$  is necessary due to sublimation of  $\text{Cr}_2\text{O}_3$ . The homogeneous mixture is fired at  $1200^\circ\text{C}$  for a period of 6 hours followed by grinding and firing at  $1200^\circ\text{C}$  for another 6 hours. The product is reground, moulded and fired at  $1600^\circ\text{C}$  for a period of 10 hours. Check by chemical analysis was necessary to ascertain that MgO and  $\text{Cr}_2\text{O}_3$  molar ratio was always 1:1.

#### *$\beta$ -Dicalcium silicate and tricalcium silicate minerals*

$\beta$ -dicalcium silicate ( $2\text{CaO} \cdot \text{SiO}_2$ ) and tricalcium silicate ( $3\text{CaO} \cdot \text{SiO}_2$ ) minerals are present in Portland cement.  $\beta$ -dicalcium silicate mineral inverts on cooling at  $675^\circ\text{C}$  to the  $\gamma$ -form. The preparation of standard  $\beta$ -dicalcium silicate was carried out by heating a mix of finely ground CaO and  $\text{SiO}_2$  in the molecular ratio 2:1 at  $1450^\circ\text{C}$ . The process of regrounding and reheating was repeated until the preparation was homogeneous. The product is then reground, moulded and fired at  $1600^\circ\text{C}$  for a period of 10 hours. The product is then cooled slowly till  $1000^\circ\text{C}$  and then quenched in air to obtain the  $\beta$ -form of dicalcium silicate.

Tricalcium silicate is stable only over a limited temperature ranging from  $1250$ – $1900^\circ\text{C}$  and decomposes both above and below this range into dicalcium silicate necessitating a series of burns. A mix of CaO and  $\text{SiO}_2$  in their molecular ratio was heated at  $1200^\circ\text{C}$  for a period of 6 hours. This process was repeated until the

combination was complete. The product was ground, moulded and fired at 1600°C for a period of 10 hours followed by quenching rapidly in air. X-ray diffraction patterns of the prepared minerals proved their complete formation.

#### 4. RESULTS AND DISCUSSIONS

The minerals are pressed in the cell under low pressures less than 100 kg/cm<sup>2</sup>. The packing density  $p$  is calculated from the specific gravity of the samples and the dimensions of the cell. The true value of the dielectric constant  $\epsilon'$  is calculated from the measured value  $\epsilon'_m$  using eq (2). Also, the attenuation ( $\alpha$ ) is calculated using eq (1). The results obtained for  $\epsilon'_m$ ,  $\epsilon'$  and  $\alpha$  are given in table 1.

Table 1. Results of the dielectric constant  $\epsilon'$  and attenuation  $\alpha$  for the investigated samples.

Substance	$r$ Å	$V$ (Å) <sup>3</sup>	Atomic weight	At. wt / $V$	Specific gravity	$P$	$\epsilon'_m$	$\epsilon'$	$\alpha_{\text{dB}}$
Chromstatite (MgO.SiO <sub>2</sub> )	5.28	630	100	0.160	3.19	0.335 0.355 0.405 0.432	1.80 1.89 2.27 2.41	3.39 3.51 4.13 2.27	0.235 -0.280 -0.400 -
Forsterite (2MgO.SiO <sub>2</sub> )	7.34	1630	140	0.012	3.22	0.458 0.461 0.474 0.497	2.37 2.46 2.52 2.68	3.99 4.17 4.21 4.37	0.240 -0.255 -0.275 -
Magnesian- chromite (MgO.Cr <sub>2</sub> O <sub>3</sub> )	7.52	1740	192	0.011	4.39	0.345 0.358 0.363	2.41 2.47 2.51	5.09 5.11 5.16	0.192 -0.203 -
Magnesian- ferite (MgO.Fe <sub>2</sub> O <sub>3</sub> )	7.54	1805	200	0.011	1.20	0.435 0.511 0.536	3.03 3.63 3.90	5.67 6.16 6.40	-0.205 -0.255 -0.280
$\beta$ -Dicalcium Silicate (2CaO.SiO <sub>2</sub> )	8.00	2150	168	0.078	3.28	0.369 0.381 0.384 0.399	2.19 2.29 2.37 2.40	4.22 4.39 4.57 4.52	-0.150 -0.270 -0.300 -0.400
Tricalcium Silicate (3CaO.SiO <sub>2</sub> )	10.39	4720	228	0.040	3.15	0.422 0.437 0.443	3.78 3.95 3.97	7.60 7.74 7.73	-0.255 -0.275 -0.300

To discuss the dielectric behaviour of these crystalline materials, it will be helpful to estimate their dimensions from the atoms and ions radii (Kleber 1970). To a first approximation, the atoms and ions can be considered as rigid spheres, so that the crystal radius can be taken as the sum of the concerned radii. For more accurate interpretation we have to take into consideration the polarization and deformation properties.

It is also worthy to find the variation of the packing density in the crystal from one mineral to another. As shown in table 1, the volumes of the crystals

are calculated using the estimated radii and the ratios of the atomic weight to the corresponding volume are given. This ratio is proportional to the packing density in the crystal and is given also in table 1.

The relation between  $\epsilon'$  and  $P$  is illustrated in Fig. 1 for the three groups investigated in this work. As expected  $\epsilon'$  increases linearly with  $P$  i.e., with the increase in pressure. Similar behaviour was found before in the case of titanate ceramics (Marks & Monson 1958). It is interesting to find that for each group, the mineral of lower specific gravity has the higher dielectric constant as the same packing density.  $\epsilon'$  for calcium compounds at any pressure are higher than the magnesium compounds. This was noticed before in the case of Mg and Ca-titanates (Marks & Monson 1958).

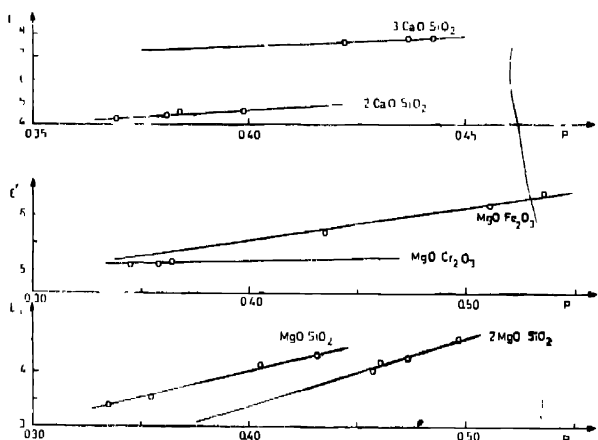


Fig. 1 Dielectric constant  $\epsilon'$  versus packing density  $P$  for the different samples

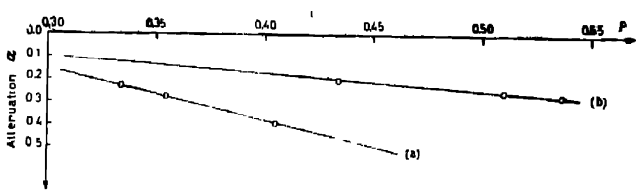


Fig. 2. Attenuation  $\alpha$  versus packing density  $P$  for  
a)  $\text{MgO} \cdot \text{SiO}_2$   
b)  $\text{MgO} \cdot \text{Fe}_2\text{O}_3$

Examples of the attenuation of the samples in decibels are shown in Fig. 2 at the different packing densities. The curves show that  $\text{MgO} \cdot \text{SiO}_2$  having the smallest radius of all the investigated magnesium minerals exhibits the highest attenuation, while  $\text{MgO} \cdot \text{Fe}_2\text{O}_3$  having the largest radius shows the lower attenuation. The attenuation of the other samples lies in between these two curves.

This may lead to the conclusion that the attenuation in this frequency range depends upon the dimensions of the crystal. Also since  $\text{Mg Fe}_2\text{O}_4$  has low attenuation and high electrical resistance (known to be between  $10^2$  and  $10^6 \Omega$  (cm)), it can be used as cores in high frequency coils. It is to be added that tetracalcium silicate possesses high dielectric constant and low attenuation as shown in table I which are the properties of a good insulator and so it could be used as an insulator.

The dielectric loss  $\epsilon''$  of the investigated materials is low and the reflection method adopted here was not suitable for its determination. A method described by Roberts & Von Hippel (1946) using two sample lengths is more convenient but it was difficult to press two samples under the sample pressure and so  $\epsilon''$  could not be measured by the method used in this work.

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